- 1 The role of plastic chemical recycling processes in a circular economy context
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## 15 ABSTRACT

It is estimated that nearly 400 million tons of plastic are produced each year worldwide. However, only 10% 16 17 of this enormous amount is recycled after use. Currently, mechanical recycling is the dominant method, despite 18 certain operational limitations. To increase recycling rates, chemical recycling processes are emerging as viable 19 alternatives, promising the creation of more valuable products. This comprehensive review begins with an 20 introduction to ongoing plastic recycling technologies, covering general pretreatment methods for plastic waste 21 and the taxonomy of various recycling technologies and their applications for specific polymer recycling. Then 22 various aspects of chemical recycling are examined to explore its role within the context of a circular economy. 23 Detailing chemical recycling technologies, such as depolymerization pathways and thermochemical pathways, are 24 systematically elaborated. It also delves into optimization strategies, technological maturity, and economic 25 assessments of chemical recycling. In addition, this review also examines the symbiotic and/or substitutional 26 relationship between conventional recycling methods and alternatives, including biological recycling, 27 biodegradable polymers, and eco-design. Finally, the approaches to improve the large-scale application of 28 chemical recycling technology from the perspectives of technological level, infrastructure construction, public 29 awareness enhancement, and national and international policy formulation are discussed. This review aims to 30 provide theoretical support and practical recommendations for the future development of chemical recycling 31 technologies to achieve the goals of sustainable development and a circular economy.

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33 Keywords: Plastic Waste; Chemical Recycling; Technology Readiness Level; Value Chain; Circular Economy

## 34 1. Introduction

Although plastics have provided functional and economic benefits to our society since the 1950s, their production and use inevitably generate waste. Based on a socio-economic projection of plastic production, it is expected that the total production will reach over 1,340 million metric tons (Mt) by 2050, which is almost three

- times compared to the production volume in 2021 (390.7 Mt) [1]. In recent years, the production of plastic materials
- 39 has shown a steady upward trend, despite widespread awareness of the environmental impacts of certain plastic
- 40 materials beyond their intended use (Fig. 1a). Based on the available data, we observed that Asia accounted for
- 41 more than half of the world's production in 2021(52%), with China alone contributing 32% to the world's plastic
- 42 production, followed by North America and Europe, which will contribute 18% and 15% of the world's plastic
- 43 production, respectively (Fig. 1b).
- 44



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Fig. 1 Global plastic production: (a) from 2018 to 2021, (b) distribution by geographic area, (c) distribution by application, and (d) distribution by polymer types, across the world in 2021 [2,3]. In Fig. 1b, CIS stands for The
Commonwealth of Independent States. In Fig. 1d, PS stands for polystyrene, PUR for polyurethanes, PP for
polypropylene, HDPE for high-density polyethylene, LDPE for low-density polyethylene, PVC for polyvinyl
chloride, and PET for polyethylene terephthalate.

51 Over the years, the plastics market has shown steady growth, with the packaging sector leading this expansion, 52 and accounting for almost half of the annual plastics production (with 44%, as shown by Fig. 1c). This growth is 53 driven by products such as polyethylene (PE), polypropylene (PP), and polyethylene terephthalate (PET), as 54 illustrated in Fig. 1d. The building and construction sectors follow closely, accounting for nearly one-fifth of the 55 market (Fig. 1c, data from 2021) [2,3]. Industrial sectors such as the automotive, electronic components, and 56 electronics industries account for 15% of the global plastics market. As another major field of plastic applications, agriculture and farming activities account for 4% of the total plastic usage. Most of these plastics are used for 57 58 agricultural purposes, such as greenhouse films, mulching films, and packaging for fertilizers, seeds, and 59 pesticides. Similar to packaging plastics, plastics used in agriculture are mostly in the form of films, which are 60 more challenging to recycle and manage compared to packaging plastics. Therefore, these agricultural plastics 61 should also attract sufficient attention.

62 However, only 8.3% of global plastic production consists of recycled plastics, and only 1.5% comes from 63 renewable resources. In simple terms, this means that a substantial 90.2% of production still depends on virgin 64 molecules derived from fossil resources, mainly oil and natural gas (**Fig. 1d**). This evidences that the incorporation 65 of recycled plastics into industrial value chains struggles to materialize within a circularity framework [4].

Although it appears to be a promising endeavor, the recycling of plastic waste involves a complex array of technical intricacies and nuanced considerations, ranging from logistics to health concerns. Currently, there are four well-defined categories for recycling end-of-life plastic materials, known as primary and secondary recycling routes (via mechanical processes), tertiary recycling (via chemical routes) and quaternary recycling (a term used for direct incineration) [5]. These four routes are further presented in sections 2 and 3.

Nowadays, mechanical approaches remain the most widely used technologies for the large-scale treatment of plastic solid waste (PSW) [6]. However, the global recycling rate remains relatively low because these recycling methods are only effective for certain specific polymer materials (mainly PET and polyolefins), and often for pure, unmixed streams. In fact, in the United States and Europe, the overall recycling rate for plastics from household and collective uses is less than 30% [4,7].

76 As a result, the accumulation of PSW reached approximately 353 Mt in 2019. While some of this PSW is 77 mechanically recycled, the majority is either incinerated and landfilled (in certain countries) or mismanaged and 78 dumped in terrestrial and aquatic ecosystems [8,9]. This release of plastic into the environment collaterally induces 79 the formation of microplastics and nanoplastics, which raises concerns about ecotoxicity. This current end-of-life 80 of PSW is illustrated in Fig. 2, which was generated using data from the Organization for Economic Cooperation 81 and Development (OECD) ENV-Linkages model [10]. While the proportion of PSW subjected to incineration 82 seems to remain constant (around 18%), its mass will undergo a pronounced amplification (increasing by almost 83 67 Mt to cross the threshold of 182 Mt). Through a ripple effect, the volume of unmanaged post-consumer waste 84 is expected to increase from 79 to 153 Mt. In addition, the dominant method of controlled landfill disposal is 85 expected to continue as the predominant modality for the management and post-consumer treatment of plastics. 86 By 2060, projections suggest that the mass of PSW landfilled will increase to 507 Mt, representing more than fifty 87 percent of plastic materials facing their final fate [10]. In the current context, with absent regulatory incentives or 88 substantiation for chemical recycling techniques, the proportion of plastics earmarked for recycling - primarily of 89 a mechanical nature - will remain in a state of inertia, hovering around the 17% threshold.



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Fig. 2 PSW management per category (mechanical/chemical recycling, incineration, landfilling and
mismanagement), in million metric tons, from the OECD ENV-Linkages model. Data illustrated for 2019 and as
a projection for 2060.

Chemical recycling remains a strategic pathway for treating specific types of PSW, including composites, unsorted streams, thermoset plastics, and plastics that have been exposed to certain pathogens or microorganisms, in a circular system. Although some chemical approaches are still either economically unviable or not fully technically feasible, according to the European Commission, adopting a circular economy model could lead to an expected reduction of around 450 Mt of carbon emissions and savings of around 600 billion Euros by 2030 [10– 13].

Chemical recycling is beginning to gain traction among researchers and those interested in implementing it.
 A wealth of comprehensive reviews of chemical recycling already exists. However, questions about its role in circular economy strategies, sustainable strategies, and economic drivers still need to be explored in depth.

103 In addition to these concerns, this study addresses other important perspectives and discussions. These include 104 the potential application scenarios, the technology readiness level, and the value chains of chemical recycling 105 methods. The risk of an imbalance between plastic waste generation and chemical recycling rates, as well as the 106 relationship between waste recycling and degradable polymers are also discussed in this work. In view of the 107 escalating environmental pollution caused by plastic waste and the ongoing issue of energy shortage, accelerating 108 the development and implementation of chemical recycling is an urgent and inevitable task. It is expected that this 109 study will provide researchers, industry stakeholders, and policy-makers with a broader perspective on chemical 110 recycling in the context of a circular economy.

## 111 2. Ongoing technologies for plastics recycling

112 In the context of advancing towards a circular economy, the recycling of plastic solid waste (PSW) is pivotal. 113 Currently, a variety of methods are employed for plastic recycling, each offering distinct advantages and limitations 114 based on the type of material and its intended application. Rather than seeking a single perfect solution, a more 115 effective approach involves combining these techniques according to the specific characteristics of the waste 116 stream. This ensures efficient management of PSW and directs the resulting products into established or emerging 117 value chains, supporting a circular economy model. This section aims to outline the current landscape of recycling 118 technologies, providing a detailed classification and overview of the alternatives available. Given that conventional 119 recycling methods may not address every type of PSW effectively, it is crucial to first address fundamental steps 120 in waste management before considering chemical recycling. This comprehensive discussion includes general 121 pretreatment processes, the taxonomy of recycling technologies, and specific recycling methods for different 122 polymers.

#### 123 2.1 General pretreatment of plastic waste before recycling processes

The critical constraint for effective recycling of post-consumer plastics is that existing recycling processes typically only accommodate sorted, single-material streams, characterized by limited external contamination (including pollutants and hazardous materials). This reality contrasts sharply with the situation for most plastics

127 found in households (where a proportion ends up in general waste streams, mixed with food leftovers and various

- types of waste in many countries), as well as plastics from medical procedures and practices [5]. For example, the
- 129 mechanical recycling of PSW with harmful additives, such as flame retardants (especially bromide-containing
- 130 molecules), stabilizers, and plasticizers could increase environmental pollution, including water contamination
- 131 [14,15]. Simultaneously, it is essential to sterilize plastic waste from the healthcare sector before subjecting it to
- 132 mechanical processes. The justification for intentional incineration of these contaminated materials is firmly rooted
- in adherence to health-centered protocols and recommendations [16].
- 134 Consequently, before any recycling option, PSW is subjected to specific pre-recycling steps, the number and 135 nature of which vary depending on the type of polymers to be recycled, the level of contamination (both in terms
- 136 of type and quantity), and the intended recycling technology to be subsequently applied [17,18]. In general, these
- 137 pre-recycling steps include: (i) collection; (ii) sorting of PSW based on its intrinsic characteristics; (iii) shredding;
- 138 and (iv) removal of impurities (Fig. 3).



139

140 Fig. 3 General steps for obtaining plastic feedstock from plastic waste for recycling technologies.

Both collection and sorting are the cornerstones of any recycling initiative. They play an important role in reducing the cost of recycling by segregating waste into different polymer classes, thereby enabling the targeted application of specific technologies [19]. Sorting of plastic streams can be performed either manually or by exploiting their physical and chemical properties. **Table 1** provides examples of different sorting techniques [20– 23].

# **Table 1.** Examples of common sorting techniques, according to some authors [20,21].

Sorting type	Property exploited	Principle
Automated optical sorting		It uses high-speed cameras and spectral sensors to recognize and distinguish various types of plastic waste by studying the form, color, light reflectance, and light absorption features of the material.

Thermal adhesion sorting	Softening point	By heating a surface, plastics soften according to their softening temperature. Consequently, some of them start adhering to the surface, allowing the segregation of divergent groups.
Float-Sink sorting	Specific gravity	Wastes are thrown into a solvent pool, whose specific gravity is adjusted to divide plastics into groups above or below their density.
Dry zig-zag sorting	Specific gravity	Plastics are placed in contact with a counter-current airflow, causing some materials to drop or rise towards corresponding exits.
Electrostatic sorting	Work function	Charged objects are subjected to an electric field and separated according to the magnitude of their charge.
Froth flotation sorting	Hydrophobicity	Similar to float-sink sorting but employing an aqueous solution of sodium sulfonate, to separate materials based on their hydrophobicity.
NIR sorting	NIR absorption	Wastes are subjected to near-infrared wavelengths, and specific absorptions are registered and used to discriminate between groups.

147 Once sorted, the subsequent shredding and extrusion steps serve to increase the effectiveness of the 148 pretreatment and recycling methods by reducing the size of the material particles. This reduction improves heat 149 and mass transfers [24]. Therefore, a range of processes are used, depending on the specific component to be 150 recovered and the recycling technology selected. Several pre-treatment methods are typically required prior to 151 chemical recycling processes, including: (i) washing; (ii) thermal dehalogenation; and (iii) solvent-based 152 purification (or dissolution). Washing consists in the introduction of cold or hot water or dilute basic aqueous 153 solutions, such as NaOH, to remove salts and inorganic contaminants. However, subsequent drying is often 154 required, particularly when certain thermochemical recycling routes are employed [17]. Thermal dehalogenation 155 is used to remove halogen compounds from plastic waste, which is particularly important in the chemical recycling 156 of polyvinyl chloride (PVC). Halogens can interfere with thermochemical processes due to their corrosive nature, 157 releasing substances such as acid gases (e.g., HCl and H<sub>2</sub>S). In addition, thermal dehalogenation assists in the 158 removal of benzene species. Typically, this process is performed at temperatures in the range of 240 to 340 °C 159 [17,24]. Higher temperatures could potentially affect the basic molecular structure of the polymer [17]. Finally, 160 solvent-based purification involves dissolving the primary polymer in the waste feedstock using a selective solvent, 161 while suspending impurities such as pigments, dyes, metals, and additives. The subsequent separation and recovery 162 of the solvent results in a waste feed with improved recyclability potential. This technique is widely used on an 163 industrial scale, particularly in the production of recycled plastic-based pellets [17,25].

# 164 2.2 Taxonomy of plastic recycling technologies

165 As noted above, plastic recycling technologies can be divided into four distinct categories, introduced primarily 166 by the American Society for Testing and Materials (ASTM) D7209 standard, namely primary, secondary, tertiary, 167 and quaternary recycling options (Fig. 4). Primary and secondary recycling routes refer to mechanical methods, in 168 which PSW is subjected to sorting, grinding, washing, and final extrusion to produce batches of recycled pellets. 169 The advantage of primary recycling is its high economic efficiency, but the disadvantage is the limited amount of 170 waste suitable for this approach, as most post-consumer waste does not meet the required specifications for primary 171 recycling. An illustrative example is the "bottle-to-bottle" recycling of only certain high-purity PET grades [5]. 172 Secondary recycling primarily involves the mechanical transformation of plastic waste into materials other 173 than those for which the original materials were intended. This process typically results in products of lower value

and quality, hence it is usually called "downcycling." Although the recycling rate is not high, a significant portion of post-consumer materials undergo downcycling. For example, plastic waste from food packaging can be granulated and repurposed into non-food-grade plastic items, such as flower pots, paint buckets, shampoo bottles, and drip irrigation tape [5]. Besides, one drawback of primary and secondary recycling is that plastics can only be recycled for a limited number of cycles due to a decrease in molecular weight and resulting lower performance.

Conversely, the intentional breakdown of polymers into repolymerizable monomers or other lower molecular
weight building blocks (for the chemical or energy sectors) under controlled conditions is referred to as "chemical
recycling" or "tertiary recycling" [5,26]. These tertiary recycling techniques are often referred to as "upcycling"
methods (See section 3).

Finally, quaternary recycling refers to incineration, where plastic waste is burned to generate heat. This heat can be captured and used to heat buildings or generate electricity. Countries such as the United States, Japan, and Singapore have adopted this approach [27,28]. However, this process raises environmental concerns, including the production of toxic gases and heavy metal-contaminated ash. The environmental and health impacts of waste incinerators are highly dependent on the emission control technology, as well as the design and operation of the incinerator [29].



## 190 Fig. 4 Taxonomy of the different PSW recycling technologies.

# 191 2.3 Recycling Methods for Specific Polymers

192 As noted above, not all PSW are suitable for all recycling technologies. Plastic materials, which are 193 characterized by different molecular structures, have intrinsic properties that vary widely. These properties include 194 density, melting temperature  $(T_m)$ , glass transition temperature  $(T_g)$ , maximum thermal decomposition temperature 195  $(T_{\text{max}})$ , and other mechanical properties. While these characteristics direct these polymers towards different 196 applications, they also influence the recycling strategy. To facilitate the proper assignment of each polymer 197 category to its appropriate recycling path and to improve sorting accuracy, specific codes ranging from 1 to 7 have 198 been introduced by the Society of the Plastics Industry (SPI). Pure PSW is identified by codes 1 to 6, while end-199 of-life mixed plastics are grouped under the SPI code 7. Table 2 (available in the supplementary material) 200 summarizes some of these properties, recycling rates and recycling methods for polymers based on their respective 201 SPI codes. A brief description of these materials, and their current recycling methods is also discussed below.

## 202 2.3.1 Polyethylene terephthalate: PET (SPI code 1)

PET stands as a resilient and malleable plastic material rendering it highly suitable for beverage containers, fibers, and filaments. Notably, PET is the predominant polymer utilized for single-use packaging on a global scale, finding application in a range of industries [30]. Given its efficient and effective collection and sorting, PET boasts the highest recovery rate among polymers on a global scale [30,31]. Typically, most PET is recycled through secondary recycling despite undergoing a decrease in chain lengths and ductility during its mechanical recovery. Nevertheless, the resultant materials remain qualified for fibers manufacture [30,32].

## 209 2.3.2 Polyolefins: HDPE, LDPE, and PP (SPI codes 2,4, and 5, respectively)

210 As the recycling approaches are similar to each other, HDPE, LDPE, and PP are usually discussed together in 211 literature [30,33–35]. HDPE is usually used in toys, utensils, films, bottles, and pipes; LDPE for packaging for 212 computer hardware, trays and general-purpose containers, cling wrap, and grocery bags; and PP serves for reusable 213 food containers, prescription bottles, bottle caps, carpets, rugs, and mats [36,37]. Like PET, mechanical approaches, 214 are used for recycling those polyolefins. Owing to its remarkable thermal stability, as well as higher crosslinking 215 rates than PP [38], polyethylene can survive melting and remolding cycles multiple times during mechanical 216 recycling processes. For example, LDPE can be extruded up to 40 times at a temperature of 240 °C [30]. Due to 217 the C-H groups at tertiary carbon centers in polypropylene being more susceptible to oxidation during reprocessing, 218 the mechanical properties of polypropylene deteriorate more rapidly when recycled through mechanical means 219 than those of polyethylene. When considering applying tertiary recycling to those polyolefins, pyrolysis appears 220 as the most efficient, although the thermal cracking is more difficult in HDPE than in LDPE and PP [39].

#### 221

#### 2.3.3 Polyvinyl chloride: PVC (SPI code 3)

PVC is an exceedingly versatile polymer, capable of giving rise to an array of products with varying lifespans, spanning from long-term applications such as water pipes and medical equipment to short-term items like raincoats, tablecloths, and shower curtains. Challengingly, in the realm of processed PVC blends, an array of functional additives is employed, enhancing the properties of PVC products or rendering them more amenable to processing. These additives include thermal stabilizers, lubricants, flow modifiers, agents for enhancing mechanical properties, plasticizers and, typically, a substantial quantity of mineral fillers, such as chalk, talc, and titanium white [40,41]. However, those additives in PVC-based materials can be leached during the recycling process, but also compromise the quality of the recycled products.

230 As PVC waste remains one of the most challenging materials to recycle, it has for a long time been incinerated. 231 However, direct incineration leads to the emission of toxic gases (SO<sub>2</sub>, NO<sub>x</sub>), heavy metals, and solid particles 232 through the flue gas. It also contributes to the formation of hazardous compounds like polychlorinated 233 dibenzodioxins and other chlorinated organic compounds [42]. Currently, secondary recycling of PVC is mastered, 234 although the intricate blend of additives found in PVC waste implies case-by-case adjustments, which make 235 secondary recycling more expensive than for PET or polyolefins [43,44]. Pyrolysis of PVC has been investigated, 236 even if it induces HCl emission and subsequent eventual equipment depreciation. A mechanochemical strategy 237 can be used to grind PVC with a HCl scavenger, such as CaO. This approach yields a calcium salt by-product that 238 can be readily removed through washing [45]. Another innovative method involves co-pyrolysis of PVC with 239 cellulose, effectively adsorbing HCl and concurrently reforming cellulose pyrolysate [46]. In essence, advancing 240 more efficient techniques, catalysts, and HCl inhibitors remains imperative to effectively recycle PVC waste 241 through chemical procedures [30].

## 242 2.3.4 Polystyrene: PS (SPI code 6)

PS is a highly versatile and chemically stable polymer, making it a popular choice from appliances and automotive parts to electronics, food services and packaging. PS is used in both solid and expanded forms. Generalpurpose PS is transparent, rigid, and rather brittle. While expanded polystyrene (EPS) foam, often called PS foam or styrofoam, is frequently used for insulation and packaging purposes. A unique practical barrier for PS recycling compared to other plastic recycling is that much of the PS waste is in the form of EPS foam, which has a high volume-to-mass ratio, thus making it more expensive to transport to a central recycling facility [37].

Besides employing dissolution techniques (which is not included in this review), PS can be broken down by pyrolysis in the presence of a magnesium-impregnated solid catalyst, resulting in a mixture of hydrocarbons such as benzene, toluene, ethylbenzene, styrene, and  $\alpha$ -methylstyrene [47]. The maximum temperature of which process is approximately 550 °C [48]. Research into the breakdown of PS in supercritical solvents (benzene, toluene, ethylbenzene) at 310-370 °C revealed that toluene was more successful than the others in recovering styrene from PS [49].

## 255 2.3.5 Miscellaneous (SPI code 7)

SPI code 7 encompasses a diverse array of thermoplastics and thermosets, distinct from those categorized under SPI codes 1 to 6. It also includes biodegradable plastics. Among these thermoplastics and thermosets, notable mentions are polycarbonate (PC), polyamides (PA), acrylonitrile butadiene styrene (ABS), polymethyl methacrylate (PMMA), polyethersulfone (PES), polytetrafluoroethylene (PTFE), polyurethanes (PUR), and epoxies, among others.1

While certain polymers from this category can be mechanically recycled, they often partially lose some of their
 physical properties. Investigations have delved into the pyrolysis of various of these "SPI 7" polymers, including
 PC, PTFE, PA, PMMA, and even PUR and epoxy composites [11,26,50–55]. Unlike PE and PP which fragmentize

- randomly, PTFE, PA and PMMA can be pyrolyzed into products primarily composed of their respective monomers
- [11]. For instance, pyrolysis of PMMA yields an impressive monomer content of nearly 98% [50]. PET, PC, and
- 266 PUR are all amenable to depolymerization through solvolysis. However, solvolysis yields a mixture of final
- 267 products (either high molecular-weight oligomeric polyols or aromatic N-containing oligomers) posing challenges
- in terms of separation and purification [26,56].
- 269

SPI code	SPIChemical nameSPIcode(Abbreviations)Identifi			s [5,25,57–59	9]				Recycling rate from	Total recycling	Current main recycling methods
	()	cation	Density (g·cm <sup>-3</sup> )	T <sub>m</sub> (°C)	<i>T</i> <sub>g</sub> (°C)	T <sub>max</sub> (°C)	$\sigma_{\max}$ (MPa)	E (GPa)	containers & packaging (%) [7]	rate in MSW (%) [7]	
1	Polyethylene terephthalate (PET)	PETE PETE	1.33-1.45	280-320	70-85	435	47	3.1	25.4	18.5	Mechanical recycling, pyrolysis, solvolysis (glycolysis, aminolysis, alcoholysis, hydrolysis) [25,30,60–62]
2	High-density polyethylene (HDPE)	HDPE	0.94-0.96	200-300	-130 100	460	18-35	0.07-1.40	14.8	8.9	Mechanical recycling, pyrolysis [39]
3	Polyvinyl chloride (PVC)	$\lambda_{3}$	1.38-1.55	180-210	80-90	290, 465**	50-75	1.0-3.5	Neg.	Neg.	Mechanical recycling, pyrolysis [5,45,46]
4	Low-density Polyethylene (LDPE)		0.91-0.93	160-260	-130 100, -30 - -10	460	8-23	0.2-0.5	9.9	4.3	Mechanical recycling, pyrolysis [39]
5	Polypropylene (PP)		0.90-0.91	230-270	-20 - 20	470 (in air)	21-37	1.1-1.3	2.7	0.6	Mechanical recycling, pyrolysis [39]
6	Polystyrene (PS)		1.05	180-280	80-105	435	45-65	3.2-3.25	3.6	0.9	Dissolution, pyrolysis, and solvolysis [47,49,63,64]
7	Others*	OTHER	varies	varies	varies	varies	varies	varies	Neg.	26.7	Mechanically recycling for PC, PES and epoxy [32,65,66], pyrolysis for PC, PTFE, PA, PMMA, PUR and epoxy composites [11,26,50–55] solvolysis (alcoholysis, glycolysis, hydrolysis, and aminolysis) for PC and PUR [26,56,67,68]

#### 270 Table 2. SPI codes, recycling rates, recycling methods for different types of plastic solid waste

271 Neg.= Negligible, less than 0.02%.

272

\* Including polylactic acid, acrylonitrile butadiene styrene, nylon and polycarbonate. \*\* There are two separate degradation regimes and relative two maximum mass loss temperature for PVC. 273

274 *T*<sub>m</sub>: Melting temperatures.

 $T_{\rm g}$ : Glass transition temperature. 275

276  $T_{\text{max}}$ : Temperatures at which mass loss rate is the highest ( $T_{\text{max}}$ ) obtained from TGA results rounded to nearest 5 °C and measured at 10 °C·min<sup>-1</sup> in N<sub>2</sub> unless stated.

 $\sigma_{\rm max}$ : Maximum tensile strength. 277

278 *E* (GPa): Tensile modulus.

# 279 3. Chemical recycling: a valuable tool for a circular economy

As stated before, applying mechanical recycling does not allow to completely valorize plastic solid waste (PSW) as materials downgrade towards applications with lesser economical value. Even though it enables the reincorporation of these materials towards the existing economy, it does not take advantage of the entire value of these waste streams. Therefore, chemical recycling emerges as a critical alternative in the pursuit of a circular economy, addressing the limitations of traditional methods.

285 In this section, we will explore the role of chemical recycling as an essential component of circularity. We 286 begin by defining chemical recycling and examining the various technologies available, along with potential 287 optimization strategies. Despite varying levels of technological maturity, we will evaluate the current status and 288 future prospects of these technologies to underscore their significance. We will then present different pathways for 289 treating PSW, including both homogeneous and mixed streams, and outline the potential value chains for recycled 290 products, acknowledging that not all PSW are equally valorized. Finally, we will provide an economic assessment 291 to identify areas for improvement in chemical recycling options, as well as to assess their socioeconomic and 292 environmental impacts.

## 293 3.1 Chemical recycling technologies

From a molecular perspective, chemical recycling involves advanced processes that convert PSW into smaller molecules - either liquid or gaseous - that can then be reused as fuels, reagents, or as building blocks for the production of new plastics. The term "chemical" entails an inherent change in the structural composition and cleavage of the major chemical bonds in the polymer structure. It is important to note that not all plastics are created equal, and different plastics undergo distinct chemical recycling processes. Polyesters (such as PET and PLA), polyamides (such as nylon 6), polycarbonate, and polyurethanes can be effectively recycled through solvolysis processes. In contrast, polyolefins (such as LDPE, HDPE, and PP) are primarily suited for pyrolysis.

Chemical recycling can be broadly categorized based on the operating temperature and the nature of the bond cleavage: either controlled depolymerization at temperatures around 300-350 °C or molecular disintegration through pyrolysis at temperatures exceeding 400 °C [24]. Controlled depolymerization involves precise cleavage of specific covalent bonds, whereas pyrolysis entails a more random destruction of the polymer structure. **Table 3** highlights the main reactions observed in chemical recycling of several different polymers treated at either a low (solvolysis) or a high temperature (pyrolysis), and provides examples of commonly recovered low-molecularweight compounds.

**308 Table 3.** Illustration of the key reactions involved in chemical recycling.

	Reactions	Equations			Ref.
<b>h</b>	Reductive		Zn(OAc) <sub>2</sub> .2H <sub>2</sub> O, PhSiH <sub>3</sub>		[69]
· ·	depolymerization	the formation of	Toluene, 110 °C, 24h	но	
So		PCL /n		1,6-hexandiol	



309 Notes: PCL: polycaprolactone; TPA: terephthalic acid; EG: ethylene glycol; BPA: bisphenol A; DMC: dimethyl carbonate;
 310 BHETA: bis(2-hydroxyethylene)terephthalamide.

311 In the following sections, we will delve into these two distinct pathways of chemical recycling: depolymerization

312 processes operating under mild conditions and thermochemical pathways characterized by high-temperature

treatments. Each pathway offers unique advantages and challenges, which will be explored in detail.

## 314 **3.1.1 Depolymerization pathways (low temperatures)**

Depending on the operating temperature, which influences the way chemical bonds within the polymer are broken, chemical recycling can be categorized into controlled depolymerization (under mild conditions) or molecular disintegration (under more intense thermochemical treatment conditions). These two pathways are described separately below.

The term "depolymerization" refers to the methodical breakdown of a polymer into its constituent monomeric units. This includes techniques such as reductive depolymerization and solvolysis [30]. Reductive depolymerization is a reaction that converts a polymer into high-value compounds, such as polyols or polyamines, through the action of a transition metal catalyst and a reducing agent (H<sub>2</sub> or silanes). Reductive depolymerization is usually carried out at relatively low temperatures, ranging from 65 to 160 °C [74]. Fernandes demonstrated that 324 it was possible to convert polycaprolactone (PCL) to hexan-1,6-diol using zinc acetate dihydrate  $(Zn(OAc)_2 \cdot 2H_2O)$ 

and phenylsilane (PhSiH<sub>3</sub>) as the catalyst and reducing agent, respectively (**Table 3**, line 1) [69]. Similarly, in

solvolysis reactions, the solvent cleaves the bonds between the monomers, with or without an additional catalyst.

327 Depending on the solvent used, different categories are distinguished, each with different resulting end products:

328 hydrolysis takes place in the presence of water, while alcoholysis employs an alcohol, often an aliphatic one.

329 Glycolysis profits from the utilization of glycols, whereas aminolysis is conducted by the action of aliphatic or

aromatic amines.

Hydrolysis has proven to be efficient for the chemical recycling of polymers containing *O*- and/or *N*-containing (co)-monomers, such as polyesters, polyurethanes (PURs), polyamides, or phenolic resins. This results in the formation of polyols, amines, or other intermediates or monomers, depending on the nature of the polymer being treated. Hydrolysis can be carried out under neutral, alkaline, or acidic conditions using hydroxides or sulfuric acid [75]. The most common example is the hydrolysis of PET at temperatures between 150 °C and 300 °C and at pressures between 1 and 4 MPa, yielding predominantly terephthalic acid (TPA) and ethylene glycol (EG) (**Table 3**, line 2) [70].

338 Alcoholysis involves the depolymerization of polymers in an alcohol-based medium, sometimes in the 339 presence of an acid or a base catalyst. The most notable approach is methanolysis using methanol. Chiu and 340 coworkers reported optimal methanolysis of polycarbonate in less than 5 min under batch conditions at 240 °C, 341 producing both bisphenol A (BPA) and dimethyl carbonate (DMC) (Table 3, line 3) [71]. Glycolysis relies on the 342 use of glycols with or without the addition of a catalyst (organometallic, amine, or acid-base catalysts) [72]. 343 Ethylene glycol (EG) is the most commonly used solvent for glycolysis and has been found to be effective for the 344 chemical recycling of PUR foams (Table 3, line 4) [72]. Finally, aminolysis uses a solvent with an aliphatic or 345 aromatic amine function, such as ethanolamine, in the presence of a catalyst such as sodium acetate or glacial 346 acetic acid. Shukla et al. [73] reported the depolymerization of PET to bis(2-hydroxyethylene) terephthalamide 347 (BHETA) with a 91% yield in the presence of ethanolamine and sodium acetate as an aminolytic agent and catalyst, 348 respectively (Table 3, line 5).

#### 349 3.1.2 Thermochemical pathways (high temperatures)

Depending on the conditions used, thermochemical processes can be categorized as pyrolysis (in an oxygendeficient atmosphere), gasification (in an oxygen-rich atmosphere) or hydrothermal liquefaction (HTL) using supercritical water (sc. H<sub>2</sub>O), as illustrated in **Table 4**.

353 During pyrolysis, the hydrocarbon chains that make up the polymers are broken down into simpler, low-354 molecular-weight units. This process generally results in the production of: (i) a gas phase, containing volatile 355 compounds; (ii) a pyrolysis oil that integrates a range of hydrocarbons, aromatics, and other liquid organic 356 compounds; and (iii) a carbonaceous residue called biochar. The relative proportions of these different fractions 357 depend on the reaction conditions applied [18,76]. Pyrolysis is typically applied to PSW at temperatures in the 358 range of 400 °C to 700 °C and under an inert atmosphere [76,77]. The minimal temperature required depends on 359 the material being treated. Polymers with high melting points typically require temperatures higher than 500 °C 360 [18]. Pyrolysis has been shown to be efficient for a wide range of plastics (PET, polyolefins, PVC, PURs, etc.), 361 but also for multilayer, multi-material, coated plastics, mixed plastic waste (MPW) and composites, eliminating, 362 to a certain extent, the need for sorting [78]. Common reactors used for PSW pyrolysis include pressure vessels, 363 tubular reactors, fixed bed reactors, and fluidized bed reactors [77]. During pyrolysis, the degradation of polymers

364 is caused by cracking and radical chain reactions. Random cleavage of molecules produces smaller alkanes, 365 alkenes, and radical chains that further react via: (i) cracking; (ii)  $\beta$ -scission; (iii) hydrogen transfer; (iv) 366 propagation; (v) disproportionation; and (vi) coupling reactions (Table 3, lines 6 to 11) [24,79]. In general, as 367 temperature increases, gas yield increases at the expense of liquid yield by accelerating reaction kinetics [18,77]. 368 Because pyrolysis is energy-intensive, catalysts are often used to reduce the activation energy required for polymer decomposition [17,24,76,77]. These catalysts can also be used to effectively remove impurities or limit the 369 370 formation of hazardous materials (e.g. chloride compounds in the case of PVC pyrolysis). Commonly used 371 catalysts include zeolites, acidic solids, heavy metal salts, and noble metal-based catalysts [17,76]. However, the 372 recovery of these catalysts is not always efficient, which increases operating costs. In addition, during in-situ 373 catalytic pyrolysis, i.e., when there is direct contact between the catalyst and the plastic matrix, material deposition 374 can rapidly deactivate the catalytic sites [17,77]. An alternative approach is ex-situ catalytic pyrolysis, which 375 involves a two-step process. This configuration has the advantage of reducing the risk of catalyst poisoning, as a 376 second catalytic stage would process the intermediate volatiles from the first reactor, facilitating their recovery and 377 reuse [24]. It should also be noted that current research is also dedicated to the possibility of pyrolyzing unsorted 378 PSW to yield certain intermediate molecules that could enable the creation of novel solvents or liquid fuels for 379 transportation purposes.

In hydrothermal liquefaction (HTL), supercritical water acts as both the solvent and catalyst. Its advantages include inhibiting undesired side reactions, improving product selectivity, removing impurities such as halogens, nitrogen, and oxygen compounds, and reducing coke/char formation. The main reactions involved are similar to those in pyrolysis, particularly cracking and  $\beta$ -scission reactions (**Table 3**, lines 6 and 7) [24]. HTL offers significant results for a wide range of PSW at temperatures ranging from 350 °C to 500 °C. However, due to the corrosive nature of supercritical H<sub>2</sub>O and the substances dragged into the gas phase under acidic conditions, stronger steel alloys are required for this process, which may increase capital expenditures [17,24].

Finally, gasification is the third thermochemical route that is efficient for recovering PSW. Operating at temperatures between 700 °C and 1000 °C, gasification aims to produce a synthetic gas, or syngas, containing mainly CO and H<sub>2</sub>. Unlike pyrolysis, gasification uses an oxidant such as air, pure oxygen, steam, or sc.H<sub>2</sub>O. Steam gasification is reported to produce a syngas with a higher heating value than air gasification due to the absence of nitrogen, resulting in hydrogen enrichment [18,76,77]. Gasification has been shown to be efficient in converting a wide range of polymers to syngas, which can subsequently be upgraded to hydrocarbon-based monomers or fuels

393 through a Fischer-Tropsch process.

394

 Table 4. Summary of the main characteristic for thermochemical recycling technologies.

Technology	Temperature range (°C)	Type of products produced	Main advantages	Particular requirements
Pyrolysis	400 ~ 700	Low molecular weight products, mixture of hydrocarbons and char.	It offers the versatility of treating mixed plastic strains, composites and/or multi-material plastics.	It demands the use and management of catalysts and various reactor configurations, increasing operational costs.
Hydrothermal liquefaction	350 ~ 500	Low molecular weight products, mixture of hydrocarbons and char.	Inhibition of side reactions, increase in selectivity and reduction of char. Mild reaction conditions.	It requires reactors made of corrosion-resistant alloys, increasing capital costs.

Gasification	700 ~ 1000	Syngas (mainly	$H_2$	A wide	range	of	Higher temperatures are
		and CO).		polymers	can	be	needed. The use of steam
				treated.			is intended for hydrogen
							enrichment.

395 3.2 Chemical recycling strategies for mixed wastes, composites and thermosetting wastes

396 The recycling strategy for MPW and composites may differ slightly from that of homogeneous waste streams 397 or functional groups that do not necessarily interfere with product quality or specific applications, especially when 398 conventional recycling techniques are used. Contamination from external substances in one type of plastic can 399 compromise the mechanical recycling of another type. For example, chlorine from PVC can pollute a polyolefin 400 recycling stream. However, submitting a mixed stream of PSW, containing both types of materials, to pyrolysis 401 does not undermine the production of pyrolysis oil. In this case, pyrolysis oil can still be produced from the blended 402 feed, but careful attention must be paid to controlling the presence of chlorine in the final product [78]. To properly 403 regulate the resulting liquid yield and quality, reaction parameters play a significant role. Different conditions can 404 not only alter the proportion of the resulting products but also affect the distribution of functional groups or 405 elements between different phases. Key parameters include the nature and ratio of the polymers, temperature, 406 heating rate, residence time, catalyst type, and reactor configuration [78,80-82].

407 On the other hand, recycling composites is more challenging, especially when thermoset plastics are used as 408 the matrix material. This is the case with some fiber-reinforced polymers (FRP). For example, the imminent 409 retirement of first-generation aircraft and wind turbines made with carbon fiber-reinforced polymer (CFRP) 410 composites over the next decade, coupled with the anticipated global CFRP waste reaching 20,000 tons by 2025, 411 underscores the critical need for sustainable recycling strategies. Common methods for managing FRP include 412 incineration (utilizing the associated heat), mechanical recycling, and landfill disposal. Although pyrolysis can 413 recover some thermally stable fibers from these composites, it also produces pyrolysis products that can affect the 414 properties of the recovered fibers due to thermal degradation. A similar issue occurs during mechanical recycling, 415 where the resulting products are downgraded. Recent advancements in recycling technology offer novel 416 approaches that promise to enhance the viability of recycling thermoset composites. Liu et al. [83] demonstrated a method for effectively recovering long carbon fibers and epoxy resin from CFRPs using deep eutectic solvents 417 418 (DESs) and metal salt catalysts (e.g., ZnCl<sub>2</sub>), achieving complete decomposition of the epoxy resin in hydrophobic 419 DESs at 180°C for 1.5 h, while the recovered carbon fibers retained 94.5% of their original strength. Furthermore, 420 Hao et al. [84] present an eco-friendly method for upcycling carbon fiber-reinforced polymer (CFRP) waste by 421 decomposing the epoxy matrix under mild conditions, converting the decomposed polymer into recyclable 422 vitrimers, and creating new hydrothermally recyclable composites with recovered carbon fibers. It is essential to 423 integrate these new recycling technologies into the automotive, wind energy, and aerospace industries to meet the 424 circular economy objectives.

Unlike thermoplastics, which are often designed for single use and require sorting before chemical recycling, certain thermosetting wastes (like tires, wind turbine blades, etc.) are bulk and easily separable from other plastic wastes. However, thermosets are covalently cross-linked polymers with exceptional mechanical strength, thermal properties, and chemical stability, making them highly desirable in structural and protective applications but particularly difficult to recycle. Mechanical reprocessing is not feasible for thermosets due to their cross-linked structure that prevents them from flowing. Chemical recycling technologies convert thermoplastics into their monomers or low molecular weight compounds, thereby enabling their reuse and becoming a crucial approach to 432 addressing this challenge. Pyrolysis has been explored for the recycling of thermoset resins [54,55]. Despite the 433 incorporation of acidic and metal catalysts, these processes are energy-intensive and produce unfavorable mixtures 434 that require purification before use [26]. Milder chemical methods have been developed to exclusively on the 435 hydrolysis of polymers carrying acid- or base-labile functional groups, such as polyurethane [85]. For epoxy resins, 436 a recent study reported a light-driven, proton-coupled electron transfer-based method for completely 437 depolymerizing insoluble thiol epoxies with varying cross-link densities and material properties [86]. This yielded 438 a well-defined mixture of monomer derivatives with a combined yield of up to 88%, which can be converted back 439 to the original bisphenol A monomer through a straightforward in situ dealkylation reaction. Furthermore, the 440 development of new thermosets has resulted in enhanced degradability by integrating stimuli-triggered cleavable 441 functional groups or dynamic covalent bonds into the network [87]. These advancements not only address the 442 recycling challenges of existing thermoset composites but also provide a foundation for designing the next 443 generation of composites that are inherently recyclable.

## 444 3.3 Optimization strategies for chemical recycling

445 Optimization of chemical recycling technologies can be based on several approaches, including improving446 recycling efficiency, adjusting the desired products, and purifying them.

The continuous processing approach can improve the efficiency of chemical recycling by reducing cycle times,
increasing throughput, and providing better control over reaction conditions, as illustrated in Fig. 5a [23].
Continuous processes can integrate multiple steps into one system, resulting in increased efficiency and costeffectiveness.

Electrochemical methods are also being explored as an alternative to improve recycling efficiency (**Fig. 5b**). These methods use electrocatalysis and electrical energy to break down polymers into valuable chemicals or monomers [88]. They offer potential advantages such as mild reaction conditions, high selectivity, and the ability to target specific polymers.

455 Catalytic hydrotreating uses a hydrogen atmosphere to remove heteroatoms (N, S, O, Cl) and metals, and to 456 reduce unsaturation and aromatics in the pyrolysis oil, resulting in a narrower product distribution. This process 457 can convert polyolefins into synthetic biofuels and recover recycled monomers from various plastics such as PET, 458 PC, PS, PPO, poly(p-phenylene oxide) (PPO), and polylactic acid (PLA). However, catalytic hydrotreating 459 requires high hydrogen pressures, which may limit its application on an industrial scale. Fixed-bed (**Fig. 5c**) and 460 fluidized-bed reactors stand out as the most common configurations [76].

461 Co-pyrolysis involves the simultaneous thermal decomposition of both plastic waste and biomass, using the 462 synergy to improve the properties of the resulting output, as illustrated in Fig. 5d. It can serve several purposes, 463 such as mitigating tar and/or char formation, adjusting gas or liquid yields, or influencing the production of a 464 specific output [28,39]. This technology offers several advantages, including the potential use of existing 465 technological infrastructure, although a fixed bed reactor is preferred. In addition, the energy requirements are 466 lower compared to those of plastic waste pyrolysis alone, as biomass degradation occurs at lower temperatures. 467 The outcome of the process is influenced not only by the compatibility between biomass and plastic, but also by 468 the ratio of biomass to plastic, an area that is currently the focus of ongoing research. Similarly, co-gasification of 469 plastic waste and biomass has been explored to reduce tar formation during the process [77].

470 Finally, distillation and fractionation, which are commonly used as the first steps in improving the purification471 of pyrolysis oil, provide an option for managing the wide range of products inherent in the pyrolysis of PSW. In

- 472 fact, distillation can efficiently separate adducts according to their respective boiling points while providing a
- 473 means to remove residual contaminants, such as metals [17,24].



475 Fig. 5 Optimization strategies for chemical recycling: (a) Continuous processing mode, (b) Electrochemical
476 depolymerization, (c) Catalytic hydrotreatment, and (d) Co-pyrolysis with biomass.

## 477 3.4 Technological maturity of chemical recycling technologies

474

478 The aforementioned chemical recycling alternatives have different technological maturity. Some are already 479 operational at industrial scale, while others are still on development, either in academic research laboratories or in 480 the private sector. Among these technologies, pyrolysis holds significant sway due to its well-established maturity 481 in the oil and gas sector [24,25,69,70]. In fact, prominent companies active in the food packaging sector are playing 482 a pivotal role in driving the advancement of these technologies [24]. They have also been prompted by waste 483 regulations worldwide, such as the European Waste Framework Directives and the United States Environmental 484 Protection Agency's National Framework for Advance Recycling [7,89]. In addition, the incorporation of recycled 485 feedstocks, such as monomers and upgraded pyrolysis oil, helps reduce the amount of fossil-based feedstocks 486 required for their processes, thereby meeting the European Union (EU) regulation of 30% recycled plastic content 487 in new polymer items [89].

To date, the emergence of new chemical recycling facilities has mostly been driven by the creation of joint ventures or supply agreements between large companies and growing start-ups or small businesses, leveraging their complementary technical expertise. This collaborative support has enabled smaller actors to secure the necessary investment for advancing their processes and ensuring economic viability. Over time, there has been a growing trend for established companies to license their technologies, leading to an increase in the number of recycling facilities expected to come online in the coming years [60,90].

494 Pyrolysis stands out as the most mature recycling technology due to its adaptability in managing mixed plastic 495 waste and the existing infrastructure and expertise. Key industry players in this area include well-known companies 496 such as Quantafuel, BASF, Neste, Plastic Energy, Shell and Sabic (refer to **Table S1** in the supplementary 497 material). The pyrolysis oil is used to produce valuable chemicals, fuels, and/or new plastics, while the char-like granules are used as raw materials for the construction and automotive industries. The gases produced during
pyrolysis are often used to generate heat and electricity, a practice that reportedly offsets the energy requirements
of the process [17,90,91].

While many operators focus on recycling mixed plastic waste to add value to non-mechanically recyclable materials, some have highlighted the challenge of ensuring consistent properties in the end products due to the diverse nature of feedstock batches. This is particularly relevant when aiming to produce high-value items from the pyrolysis oil. To address this issue, many companies have adopted a mass balance approach. This involves using the resulting oil to reduce their reliance on fossil-based crude oil consumption, providing them with a wider range of operational flexibility [60].

507 On the other hand, depolymerization routes are gaining momentum on an industrial scale, with notable players 508 such as Revalyu. Revalyu operates a recycling facility in Nashik, India, where they produce recycled polyester 509 yarns from PET waste [25,60]. This dynamic shift toward controlled depolymerization has also attracted the 510 attention of other industry leaders. Companies such as Aquafil, Ioniqa, and Eastman Chemical have successfully 511 validated their technologies up to technology readiness level 7, with validation and construction projects underway 512 for full industrialization soon (refer to **Table S2** in the supplementary material). Among the various solvolysis 513 pathways, glycolysis takes the lead due to its versatility and favorable operating cost profile [25,60].

514 Conversely, gasification and hydrothermal liquefaction (HTL) represent a limited number of systems that have 515 successfully demonstrated their capabilities at the industrial level (listed in **Table S3** in the supplementary 516 material). Prominent players in this field include Klean Industries, Mura Technologies, and Renasci, each with 517 established plants, while other projects are actively pursuing qualification. Notably, the growing interest in 518 sustainable aviation fuels (SAF) and green plastics, coupled with the need to diversify supply chains, is acting as 519 a catalyst to accelerate the development of these technologies [92,93].

## 520 3.5 Typical value chains of chemical recycling

In scenarios where mechanical recycling of plastic waste might be cost-prohibitive or unviable, chemical recycling emerges as a solution that rejuvenates plastic waste and positions it at the beginning of new value chains [94]. **Fig. 6** shows a diagram delineating the value chains associated with the main chemical recycling methods: solvolysis, pyrolysis, and gasification. This diagram highlights the resulting value-added products and their multiple applications, including the recovery of the original polymers or the synthesis of alternative chemicals.

526 Nowadays, pyrolysis is emerging as an important method for converting plastic waste into either fuels or 527 valuable chemicals. For example, pyrolysis can be used to recover the monomers of polystyrene (PS) and 528 poly(methyl methacrylate) (PMMA), namely styrene and methyl methacrylate (MMA), which can then be reused 529 in the synthesis of the original polymers. In contrast, PE and PP tend to undergo random fragmentation, resulting 530 in a wide and skewed distribution of products, constituting the pyrolysis oil [11].

531 Compared to pyrolysis, solvolysis offers a more precise approach to the recovery of polyesters and polyamides.
532 Solvolysis processes are highly effective in treating polymers containing ester, ether, and acid amide bonds, such
533 as PET, PU, PC, and nylon, among others. PET, for example, can be subjected to alcoholysis, glycolysis, and
534 hydrolysis to break it down into its constituent monomers - dimethyl terephthalate (DMT), bis(2-hydroxyethyl)
535 terephthalate (BHET), and TPA, respectively. Repolymerization of PET can be achieved using BHET and DMT
536 without conversion, although TPA must first be converted to BHET. It should be noted that hydrolysis processes
537 are preferred over alcoholysis processes [25]. It is important to note that the products derived from the glycolysis

538 of PET are of particular value because they contain oligomers (e.g., oligoesters) that can be used in the synthesis

of PUR or unsaturated polyester resins (UPR) [95,96].

540 PA6, a polymer formed by ring-opening polymerization of  $\varepsilon$ -caprolactam, can be depolymerized by hydrolysis 541 in both sub- and supercritical water conditions to yield  $\varepsilon$ -caprolactam and  $\varepsilon$ -aminocaproic acid [97]. Similarly, 542 polycarbonates (PC) can be depolymerized to BPA, which can be used to produce PC, as well as epoxy and 543 polysulfone (PSU) resins [98]. When PUR solvolysis is compared to PET, the former typically produces other 544 substances that are less suitable for reproducing the original PUR polymer. However, these byproducts can still be 545 used in the production of lower-grade PUR variants or different types of PURs altogether [99,100]. The main 546 output of the gasification process is syngas, a combination of hydrogen and carbon monoxide that holds significant 547 value. This syngas has many applications and can be used to produce a range of products, including methanol.



548

Fig. 6 Main products (in blue) and other products (in grey) obtained through different chemical recycling
approaches (in yellow) from some typical recyclable plastics (in pink), as well as more valuable products (in green)
obtained from them and the synthesis back to the original or similar polymers with them through solvolysis,
pyrolysis and gasification.

### 553 **3.6 Economic assessment of some chemical recycling options**

554 Due to certain differences in technology readiness level, conducting a comprehensive cost analysis for all the 555 different options in chemical recycling is very challenging. In addition, some quantitative data need to be adjusted 556 based on reactor configurations, individual company specifics, potential synergies among partners, the nature of 557 the post-consumer plastic waste streams to be treated, and other factors. For low technology readiness level 558 technologies for plastic waste recycling, the data available for such an analysis is very scarce. This scarcity is 559 likely attributed to the early stage of these technologies, where research and development efforts are predominantly 560 concentrated on refining the technology itself rather than on economic evaluations. Consequently, an exhaustive 561 lifecycle assessment of these emerging technologies falls outside the scope of this article. However, we present 562 here the economic analysis of high-temperature chemical recycling, namely pyrolysis and gasification, due to their 563 high technology readiness level and extensive industrial experience with these technologies. Economic 564 assessments, based on data available in the literature, are summarized in Table 5 for both pyrolysis and gasification 565 of PSW in different configurations. Four major economic indicators are reported, including net present value 566 (NPV), return on investment (ROI), internal rate of return (IRR), and payback period (PP). Therefore, NPV usually 567 depends on the selling price of the products and the electricity sales associated with the industrial-scale waste 568 plastic treatment facility. In addition, plastic recycling requires many workers for the process of plastic separation, 569 which could increase labor costs [78,101]. In general, if the NPV is negative, it indicates that the project is not 570 economically feasible.

<b>Table 5.</b> The summary of studies for economic assessment of chemical	recycling.
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Technique	NPV	ROI (%)	IRR (%)	PT (Year)	Ref.
Plasma gasification	\$ 3 M	32.1	27	5.3	[102]
Pyrolysis	-	14.2~29.5	-	-	[103]
Pyrolysis	\$ -18~28 M	-	-	-	[101]
Upcycling into porous carbon	€ 19~48 M	-	68~152	-	[104]
Gasification	€ -160~440 M	-	-	9~24	[105]
Pyrolysis (naphtha)	€ 4.3 M	-	15.7	9.2	[106]
Pyrolysis (naphtha-wax)	€ 56.5 M	-	21.3	7.3	[106]
Pyrolysis	€ 1.6 M	-	20	-	[107]

572 Notes: NPV (net present value); ROI (return of investment); IRR (Internal rate of return); PT (Payback period); M: millions. 573 There are plenty of factors that influence the profitability of plastic recycling projects. First, the profitability 574 of chemical recycling is highly dependent on the scale of operation [105]. Compared to smaller scales, large scales 575 tend to be more economically feasible due to economies of scale [108]. For example, the NPVs of pyrolysis were 576 estimated at different scales (30, 60, and 100 tons of plastic waste per day respectively). The results showed that 577 the NPV can be significantly improved by processing larger amounts of PSW, from -6.3 to 28.6 million US dollars 578 (USD) [101,108]. Secondly, the inclusion or exclusion of feedstock costs will directly affect the profitability of 579 chemical recycling projects. In most cases, the feedstock for chemical recycling is derived from municipal or 580 industrial solid waste. Therefore, some pyrolysis plants do not have to pay for the costs of feedstock procuring, 581 and may even receive additional revenue from waste disposal fees. However, on the contrary, due to the 582 competitive use of plastic waste, some pyrolysis plants need to pay for feedstock procurement, which could turn 583 the NPV defective even under the large scale of operation (-18.1, -8.6, -3.3 million USD at three scales: 30, 60, 584 and 100 tons per day) [102]. Third, the price of the final product(s) will affect the profitability of chemical recycling 585 technologies. Chemical recycling can convert plastic waste into a range of energy products. Therefore, as the price 586 of the product increases, the profitability of the plastic-to-energy project may also increase. Instead of producing 587 only naphtha, the co-production of naphtha and waxes from the pyrolysis of plastic waste can become more 588 lucrative (NPV increases from 4.3 M€ to 56.5 M€) due to the rising price of waxes [106]. Similarly, Riedewald et 589 al. [107] (2021) estimate that if the price of pyrolysis oil increases from  $\notin$  210/t to  $\notin$  227/t, the IRR of a pyrolysis 590 plant can simultaneously increase from 20% to 37%. Thus, high oil prices can stimulate the plastic-to-energy 591 business model. Finally, chemical recycling will become dependent on carbon-trading mechanisms. In general, the 592 greenhouse gases emissions from chemical recycling are significantly lower than those from conventional

incineration [105]. In addition, the greenhouse gases emissions from chemical recycling can be further reduced by
 integrating CO<sub>2</sub> capture technology, converting it to porous carbon material [103]. Therefore, by introducing a
 carbon trading mechanism and by rewarding carbon credits or imposing a carbon tax, chemical recycling can be
 beneficial for achieving carbon mitigation and neutrality goals in the long term.

597 There are still many deficiencies in the economic assessment of plastic chemical recycling. Although 598 significant progress has been made in validating the technical feasibility, environmental impact, and economic 599 viability of the other stages, comparatively little economic assessment work has been done to evaluate the recovery 600 and preparation activities [109] This is limited by the number of ongoing commercial projects, resulting in 601 insufficient information for comparison [91]. Nonetheless, this does not diminish the potential impact of 602 chemically recycled plastics on the economy and society. In the United States and Canada alone, integrating 603 recycled plastics could address a \$120 billion market, yet they currently meet only about 6% of the demand. This 604 figure only includes plastics re-entering the value chain through technologies like depolymerization or solvolysis, 605 not other outputs from thermochemical processes such as syngas, liquid fuels, or refined oils for high-value 606 chemicals [110].

Following this, the enhancement and expansion of the chemical recycling sectors would be accompanied by a stronger and more effective collection system, leading to a decrease in recycling expenses. According to the European Union, this could result in savings of hundreds of euros per ton of plastic waste collected [111]. Additionally, there would be positive impacts on society such as lower  $CO_2$  emissions and reduced reliance on fossil fuels. By increasing the use of feedstocks with more consistent prices, the sustainability of an economy built on circularity would be improved [13].

Finally, the environmental impact of plastic waste could be evaluated from the optic of any waste management 613 614 activity. On the one hand, it renders the service of treating a waste, that otherwise will be disposed in a landfill, 615 and on the other hand, it returns a product or an energy source. Therefore, the assessment is principally focused on 616 the numerous impacts that these two activities generate, both positives and negatives [112–114]. For a complete 617 life-cycle assessment (LCA) of PSW, the framework for comparison must first be established, which requires the 618 definition of the value chain to be analyzed, i.e. the feedstock to be treated (MPW, composites, or rather 619 homogeneous streams of polyolefins, PS, PVC, etc.), the technology selected for valorizing such waste (like 620 pyrolysis, solvolysis, gasification, etc.) and the selected application for the recycled good. For the latter, two 621 scenarios can be distinguished: either recycling goods enter the same value chain, i.e. they are used to produce the 622 same original polymer (closed-loop recycling), or they integrate new value chains, yielding different products 623 (open-loop recycling) [16,112,113,115] In any case, such an analysis would require performing extensive 624 calculations and analysis that are out of the scope of this work. However, interested reader may refer to the existing 625 literature on the subject.

# 626 4. Other alternatives to conventional recycling strategies

627 Despite the numerous advantages that chemical recycling provides to manage plastic solid waste (PSW), it is 628 not presented here as a flawless or unique solution. Many chemical recycling techniques are still evolving, 629 requiring advanced technology and expertise, and may not yet ensure adequate recycling rates to meet the 630 economic demands for their sustainability. This section explores alternative approaches, including biological 631 recycling and the development of biodegradable plastics, which complement or serve as alternatives to

- 632 conventional methods. Additionally, eco-design is highlighted as a crucial strategy to enhance the recyclability of
- 633 PSW, potentially improving recycling rates. Finally, a relation and comparative analysis of these approaches is
- 634 provided to elucidate the role of chemical recycling within the broader context of a circular economy.

## 635 4.1 Biological recycling

636 Two primary biological approaches have been commonly investigated to reduce the environmental impact of 637 plastic waste: (i) the application of specific microorganisms and enzymes for biodegradation of plastic waste and 638 (ii) the development of biodegradable plastics. Microbial and enzymatic degradation of plastic waste is a promising 639 strategy to depolymerize these materials into their monomers for recycling, to convert them into carbon dioxide, 640 water, and new biomass, or to produce high-value products. Enzymes capable of degrading plastics have been 641 isolated from a variety of sources, ranging from marine ecosystems and soil to plastic landfills and invertebrates 642 [116]. A number of enzymes have been identified for the degradation of PE, PS, PP, PVC, PET, and PU [117]. 643 The most extensive research among them relates to the development of exceptional PET hydrolytic enzymes for 644 PET biological cycling [118–121]. In 2016, Yoshida et al. reported a novel bacterium, Ideonella sakaiensis 201-645 F6, which efficiently employs PET as the main carbon and energy source for cell growth [118]. PETase isolated 646 from Ideonella sakaiensis shows great potential in PET degradation due to its relatively high activity at room 647 temperature compared to the other PET-degrading enzymes [119]. By determining the tertiary structures of PETase 648 and its ligand-bound complexes, Han et al. [120] elucidated the molecular mechanism of enzymatic PET 649 hydrolysis. Based on the structural information and biochemical studies of PETase, the variant PETase R280A 650 with improved PET-degrading activity was constructed [121]. Alper et al. [122] used a structure-based deep 651 learning algorithm to develop a highly robust and active PET hydrolase that demonstrated superior PET-hydrolytic 652 activity compared to both wild-type and engineered alternatives. This PET hydrolase was able to completely 653 degrade whole, untreated, post-consumer PET from 51 different plastic products within one week and as quickly 654 as 24 hours at 50 °C. Future research may focus on investigating the impact of higher modification ratios on enzyme 655 performance through the implementation of more efficient conjugation methods. Despite significant recent 656 advances in biological recycling technology, there is still room for improvement before achieving commercial 657 viability. Nevertheless, the technology has broad development prospects due to its low energy consumption and 658 environmental friendliness.

# 659 4.2 Biodegradable polymers

660 As another biological approach to reduce the environmental impact of plastic waste, the development of 661 biodegradable plastic materials has gained considerable attention. According to European Bioplastics (EUBP), 662 poly-(butylene adipate-co-terephthalate) (PBAT) and poly (lactic acid) (PLA) have become the two major 663 biodegradable plastics with the highest production capacity in the world, accounting for 4.5 % and 20.7 % of the 664 total bioplastic production capacity (EUBP, 2022) [123]. These biodegradable and bio-based plastics are designed 665 to undergo fragmentation and assimilation by microorganisms, using different conditions according to their unique 666 biodegradation characteristics. This property is advantageous in mitigating the effects of improper disposal in both 667 marine and terrestrial ecosystems, reducing contamination and accumulation of microplastics in the environment 668 [124,125]. Despite some reports claiming that biodegradable plastics may produce more microplastics, a growing 669 body of research suggests that degradation of non-biodegradable plastics may result in the persistence of microplastics [126,127]. While fragments and microplastics derived from inherently biodegradable materials aremore likely to undergo further biodegradation and eventual mineralization [124,125,128].

672 However, it is important to note that biodegradable plastics can have limitations. They may have inferior 673 barriers and mechanical properties compared to non-degradable plastic materials [129-131]. Furthermore, their 674 degradation often requires specific environmental conditions, and they may not be suitable for composting. For 675 instance, PLA is biodegradable but is not considered suitable for home composting [131]. In addition, certain 676 biodegradable plastics may not be fully biodegradable in marine ecosystems, highlighting potential complexities 677 in their overall environmental impact. Basically, given the current state of knowledge, it is not yet feasible to 678 switch exclusively to these biodegradable plastics. Nevertheless, these materials have valuable potential in specific 679 areas such as biomedical applications, packaging, and agricultural mulching. Therefore, further research and 680 development are necessary to verify the safety and applicability of biodegradable plastics prior to their widespread 681 use in the natural environment.

## 682 4.3 Eco-design

Another strategy to place PSW in a circular economy is the eco-design of the original plastic materials. Ecodesign can be described as a set of principles aimed at reducing the environmental impact of a given product throughout its life cycle without compromising its properties and functionalities. This proactive approach can be used to minimize the waste associated with materials, improve their recyclability, and overall, reintegrate the resources already in the consumption-production cycle instead of extracting and using new ones [132,133].

688 In this regard, two types of strategies are considered during the conceptual phase of product development, 689 focusing either on the material components or the material itself [132,134]. The former involves designing a 690 product to be durable, reusable, and easily integrated into a new life cycle. In the case of plastics, the choice of 691 bio-based matrices or additives would also reduce the environmental impact, although they are not always 692 biodegradable [134–136]. For example, it is possible to produce bio-based films from biomass instead of fossil 693 resources, which are mainly considered for food packaging. The eco-design of these products involves selecting 694 the appropriate combination of matrices, plasticizers, and other additives, to ensure that the material possesses 695 desirable properties while also being more environmentally friendly [136]. PLA filament for 3D printing 696 applications, hydrogels made from starch, and tablets for drug formulation made from cellulose derivatives, are all 697 examples of this approach [137–139].

698 On the other hand, the design could also focus on improving the recyclability of the product by choosing simple 699 and mono-materials or reusing recycled goods to manufacture new products. In the specific case of PSW, there is 700 currently a technical gap in the redesign of most of the non-mechanically recyclable plastics [134]. This barrier to 701 the circular economy model could be overcome by taking advantage of chemical recycling, whose main strength, 702 as already discussed, is the possibility of treating multi-layer, multi-material plastics. The role of eco-design should 703 then focus on overcoming the technical difficulties of the various chemical recycling technologies. This requires a 704 close relationship between designers and chemical recycling actors. This will lead to an increased flow of PSW 705 suitable for recycling, which is necessary for the profitability of these processes on a large scale, but also to ensure 706 a market for these new products.

## 707 4.4 Relationship and comparation among strategies

In summary, biological recycling, biodegradable polymer substitution, and eco-design are all approaches of plastic pollution management. They serve as supplementary or alternative technologies to mechanical recycling and chemical recycling technologies. Biological recycling technology for PSW mainly refers to the use of microorganisms or enzymes to recycle PSW. Biodegradable polymers are an effective way to reduce environmental pollution caused by PSW from the original source, as they serve as alternatives to recycling methods. Eco-design involves the use of recyclable plastics and/or biodegradable plastics and other degradable materials to enhance the recyclability of products from the design stage. The relationship between these three approaches and

the mechanical and chemical recycling of PSW is illustrated in **Fig. 7**.







718 The main characteristics of mechanical, chemical and biological recycling processes are outlined in Table 6. 719 In general, mechanical recycling requires a lower level of expertise, energy input, and operating costs, whereas 720 chemical and biological recycling yield a wider range of products with specific applications [140-143]. In terms 721 of environmental assessment, mechanical and biological recycling have been found to be more environmentally 722 beneficial than chemical recycling [144,145]. In addition, it should be noted that mechanical recycling does not 723 result in the production of non-CO<sub>2</sub> greenhouse gases, and biological recycling produces limited amounts of them. 724 However, certain chemical processes, such as pyrolysis, can produce non-CO<sub>2</sub> gases such as methane, ethane, or 725 other hydrocarbons [12]. Conversely, the use of pyrolysis oil, a flagship product of chemical recycling, can reduce 726 the consumption of fossil crude oil for manufacturing virgin plastics, which is not possible with other alternatives 727 [60]. In term of energy consumption, if chemical processes require more energy, it could be recovered for energy 728 generation. For example, steam and process water could be reused to reduce the overall environmental impact 729 [143].

**Table 6.** Comparison of the key aspects for mechanical, chemical and biological recycling.

Parameters	Mechanical	Chemical recycling	Biological recycling	Ref.
	recycling			
General	Well-known technology. Profit-making technique. Low level of expertise demand. Wide Industrial large-scale utilization.	Some processes are well- known while others need further development. Accomplishment of the sustainable development goals. High level of expertise demand. Industrial deployment is moderate but growing.	Need for process development. Mild conditions. No secondary pollution. Could be applied on a large scale. Lack of high-performance degrading bacteria/enzymes. Low yield of large-scale preparation of bacteria/enzymes	[11,116,142,1 46,147]
Input	Limited to monolayer plastics. Requires pre- treatment.	Efficient for diverse wastes: heterogeneous mixtures, or plastic containing halogenated compounds. Needs high quantities to be profitable. Requires pre-treatment, but fewer sorting than mechanical recycling.	Currently not suitable for all types of materials. Pretreatment of wastes to more enzyme-attackable is necessary.	[11,146,148,1 49]
Output	Excellence of products depends on the condition of input.	Specific, but pure value- added products.	Value-added products Others, including microbial biomass, salt, and CO <sub>2</sub> .	[11,117,146,1 50]
Environmental impacts	Low carbon emissions. No emissions of non-CO <sub>2</sub> greenhouse gases.	Low carbon emissions: reduce fossil fuel extraction and therefore $CO_2$ emissions. Some processes produced methane, ethane, and other hydrocarbons.	Low greenhouse gas emissions. Eventually no harmful gases and deadly compounds released.	[12,145]
Energy consumption	Low energy requirement: Most energy is used for mixing, shredding, etc. Energy cannot be retrieved.	High energy requirement: used for thermal degradation, maintaining high temperature and pressure. Energy can be recovered through energy generation of specific products.	Low energy consumption. Energy cannot be retrieved.	[12,151]

731 Despite the weaknesses and challenges that chemical recycling technologies still have to overcome, it is clear 732 that they offer a recycling solution that other alternatives do not, especially in the context of a circular economy. 733 The authors agree that the best solution to the plastic waste dilemma is first and foremost to reduce the consumption 734 of these materials, as the best recycling is the one that does not need to be performed. Secondly, improving the 735 design of plastic materials would increase the flow of plastic that can be recycled or redirected to existing value 736 chains. However, this will not eliminate the presence of plastic waste, which must be recycled to avoid polluting 737 ecosystems. For those streams of composite materials, unsorted waste, and those with varying degrees of 738 contamination that are difficult to recycle using simpler methods, chemical recycling is the solution for 739 reintroducing them into the circular economy.

## 740 5. Conclusions and recommendations

741 The changes in national and international policies, such as import and export bans on plastic waste, as well as 742 regulations on quantity and quality control of plastic waste, provide an opportunity to achieve a circular economy. This is an important step towards ending conventional plastic waste management. Chemical recycling is a powerful technique for managing plastic waste. It is a valuable solution to solve the problems of heterogeneity, complex constitution, and contamination that prevent effective and sustainable recycling of these residues through mechanical recycling. They can upcycle plastic wastes into the beginning of new value chains, or into new valuable products by using selective processes such as solvolysis or more flexible solutions like thermochemical conversions. Consequently, the demand for a circular economy has driven the innovation of chemical recycling solutions for plastic waste.

750 Except chemical recycling, biodegradable polymers and design-for-recycling polymers are gaining increasing 751 attention. Are these materials the key to solving the plastic pollution problem? The answer may be "No". 752 Biodegradable plastics are necessary in certain areas, such as agricultural mulching films, biomedicine, and 753 compostable waste bags, it is still essential to ensure the effectiveness of plastic waste collection and recycling in 754 other applications. This is because even plastics that may degrade rapidly in composting conditions may not readily 755 degrade in natural environments such as seawater or soil, let alone almost all the modifications, additives and 756 blends for the plastics that actually affect their degradability. Therefore, further research is needed to evaluate their 757 environmental behavior and effects. In terms of design-for-recycling, it is neither easy to design polymers with 758 both high recyclability and excellent properties, nor to develop catalysts with high efficiency, selectivity, tolerance, 759 and low cost to overcome the challenges of energy-intensive and unselective, as well as harsh reaction conditions 760 during recycling processes [152]. Therefore, it is difficult to achieve industrialization for designed polymers to 761 date, although most chemical recycling approaches for common commodity plastics are also far from industrial 762 application. Accordingly, a two-pronged strategy for sustainability will be pursued in the near future. In other words, the status of chemical recycling in the field of plastic pollution management is difficult to be replaced by 763 764 biodegradable polymers and design-for-recycling polymers in a short period of time.

Currently, the development levels of different chemical recycling technologies also vary. Some of them, such as pyrolysis, are already implemented on an industrial scale and their global capacity is continuously increasing. While others still need to overcome some technological limitations to become realistic alternatives to be introduced into a circular economy system. Overall, the high cost, high energy consumption, low efficiency, as well as the complicated machinery and technology involved in chemical recycling processes pose barriers to their extensive use. Many factors influence the plastics chemical recycling, and to meet these challenges, some specific scientific research, technological development and policy-making directions and can be explored:

Firstly, optimizing the waste collection system by expanding and integrating the collection of various types of waste can be helpful. Future research should focus on advancing waste sorting technologies and smart waste management systems. This includes developing more effective methods for separating different waste, such as textiles, bio-waste, and hazardous household waste, as well as implementing IoT-based solutions for monitoring and optimizing waste collection processes. Enhanced data management tools will also be crucial for tracking waste flows and informing better recycling practices.

778 Secondly, efficient and profitable chemical treatment technologies are required and need to be further developed 779 in the industry. Scientific research should focus on optimizing chemical recycling processes to enhance their 780 efficiency and reduce costs. This includes developing advanced catalysts and improving reaction conditions to 781 increase the yield and purity of recycled materials. Additionally, research should aim at minimizing energy 782 consumption and waste generation during chemical recycling. Matching technology development should emphasize scalable, affordable recycling machinery and incorporate automation and digital advancements to
enhance efficiency. Policymakers should consider implementing circular economy models that promote product
design for durability, repairability, and recyclability from the outset. This includes encouraging take-back schemes
for products at the end of their life.

787 Thirdly, to improve the effectiveness of life cycle assessment (LCA) of plastic chemical recycling, it is crucial 788 to enhance assessment tools, improve data collection, and establish standardized methods. While LCA for chemical 789 recycling has started to take a more holistic view by considering all stages of the product lifecycle, there are still 790 gaps in research that need to be addressed. A comprehensive approach to LCA can provide valuable insights into 791 the sustainability of chemical recycling processes. Efforts should be directed towards improving data collection 792 and reporting standards, particularly in terms of energy consumption, emissions, and by-products, to reduce 793 uncertainties in LCA results. Standardized methodologies for LCA in chemical recycling are essential to ensure 794 consistency and comparability across studies.

795 Fourthly, both developed and developing countries can enhance their ability and standards for sustainable and 796 responsible management of plastic waste. Governments should implement regulations and standards for managing 797 plastic waste. They should create comprehensive technical specifications and standards to ensure the safety, 798 efficiency, and eco-friendliness of chemical recycling processes. These standards should cover areas such as 799 pretreatment of recyclable materials, chemical processing, and quality control of products. Standardizing processes 800 will improve the reliability and consistency of the technology and promote uniformity within the industry. 801 Governments should also establish and enforce effective waste management policies that prioritize reducing, 802 segregating, and recycling plastic waste. These policies could include incentives for recycling facilities, 803 requirements for extended producer responsibility (EPR), and penalties for non-compliance. Additionally, these 804 policies should be customized to address the unique needs and capabilities of different regions, ensuring that both 805 developed and developing countries have the necessary frameworks to manage plastic waste efficiently.

806 Eventually, the demand for recycled plastic should be encouraged by raising consumer awareness about the 807 need and benefits of choosing recycled plastic over virgin plastic. There is a risk of a mismatch between production 808 and consumption due to the higher cost of recycled plastic products, which can be a barrier for the majority of the 809 population. Driving the demand for products containing recycled plastic would stimulate investment in chemical 810 recycling technologies, thereby increasing profitability and production of plastics with high recycling rates. 811 Policymaking should involve creating incentives that make recycled plastic products more affordable and 812 appealing. For example, policies could include subsidies or tax breaks for manufacturers that use recycled materials, 813 which can lower costs for consumers. Additionally, implementing labeling schemes that clearly identify products

814 made from recycled plastics can help consumers make informed choices.

## 815 Author contributions

816 The manuscript was written through the contributions of all authors.

# 817 Conflicts of interest

818 There are no conflicts to declare.

## 819 Acknowledgements

- 820 This work was supported by the National Key Research and Development Program of China (grant number:
- 821 2023YFD1700904), the NSFC-FNRS Joint Program BIOAGRIFILM (FNRS PINT-BILAT-M 2022, grant number:
- 42211530566), and the National Natural Science Foundation of China (grant number: 42007312).

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